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(54) EMBOSSED POLYESTER FILM PREPARATION

(71) We, E. I. Du Pont De Nemours and Company, a Corporation organised and existing under the laws of the State of Delaware, United States of America, located at Wilmington, State of Delaware, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us and the method by which it is to be performed to be particularly described in and by the following statement:—

This invention relates to basically oriented polyester film, and to a method of making it.

Oriented polyester films must have winding characteristics that permit the high speed production of uniform, stable rolls. A method previously used has been to impart surface roughness by addition of filler to the polymer blend, resulting in asperities on the surfaces of the finished film product. However, for certain specialized uses of polyester film such as magnetic recording tape, use of such a method would create roughness on both sides of the film product that would interfere with the quality of the magnetic recording. Accordingly, attempts have been made to provide a roughness on only one surface of the finished film product, so as to leave one surface of the film relatively smooth. Techniques for imparting a roughness to one surface of the film have included brushing or other post-treatment of the finished film or the extrusion of the film through a hopper die having one irregular lip. None of these techniques, however, has provided a commercially acceptable process which yields films having satisfactory winding characteristics combined with excellent recording magnetic performance.

According to the present invention, there is provided biaxially oriented polyester film adapted by surface roughness for rapid winding into uniform stable rolls characterized by a multiplicity of transverse direction ridges on one surface of the film, said ridges having a length to width ratio of greater than 1:1 and a peak-to-valley height of at least 10 microinches (0.25 micron).

The present invention also provides a process for the preparation of biaxially oriented polyester film which comprises embossing protuberances onto one surface of the film after machine direction stretch orienting, and before transverse direction stretch orienting, by

(a) heating the film to a temperature range R, between the second order transition temperature and about 40 centigrade degrees above the second order transition temperature,

(b) bringing the film into contact with an embossing surface having a surface roughness of from 30 to 500 microinches (0.76 to 12.7 microns), A.A. (arithmetic average), and at a pressure of at least about 50 pounds per lineal inch (9.0 kg/cm), the embossing surface being heated to provide a film surface temperature of at least temperature range R, and

(c) converting the protuberances into transverse ridges by transverse direction stretch orienting the film.

Apparatus which can be used in the process of the instant invention includes any conventional orientation mechanism in which an extruded film is first oriented in the machine direction and subsequently in the transverse direction. A typical apparatus which can be used is described in detail in United States Patent 2,823,421.

A machine direction orientation mechanism particularly preferred for use in the instant invention is described in United States Patent 2,547,763. The method described therein involves heating the extruded film to within the orientation tem-

perature range and stretching it between two sets of nip rolls, operating at fast and slow speeds, respectively.

While the embossing according to the present invention can be effected at any point between the end of the machine direction orientation and the beginning of the transverse direction orientation, it has been found particularly desirable, from the standpoint of apparatus efficiency, to use as the embossing surface one of the fast nip rolls of an orientation apparatus for the type described in aforementioned United States Patent 2,547,763.

The instant invention is applicable to polyester films including, for example, polyethylene-2,6-naphthalate, polyethylene-1,5-naphthalate, polyethylene terephthalate, and copolymers thereof containing up to about 5 mole percent of other ester-forming units such as diethylene glycol, other polymethylene glycols having 1 to 10 carbon atoms, hexahydro-p-xylylene glycol, other aromatic dicarboxylic acids such as isophthalic acid, bibenzoic acid and p-terphenyl-4,4′-dicarboxylic acid, cycloaliphatic acids such as hexahydroterephthalic acid, or small amounts of aliphatic acids, such as adipic acid, or a hydroxy acid such as hydroxyacetic acid.

The second order transition temperature of a particular film can be determined by methods known to the art. United States Patents 2,578,899 and 2,968,065 disclose a method for determining the second order transition temperature and representative temperatures for several thermoplastic films.

It has been found that temperatures higher than about 40 centigrade degrees above the second order transition temperature often render the film so fluid that it adheres to the surface of the nip or embossing rolls and higher temperatures also result in excessive crystallinity in the polymer film which interferes with subsequent transverse direction orientation. Temperatures below the second order transition temperature do not permit the permanent deformation of the one way oriented film. It is preferred that the temperature of the bulk of the film, at the middle of the embossing nip, be greater than about 20 centigrade degrees higher than the second order transition temperature. For most polyethylene terephthalate films, a film temperature at the center of the embossing nip of about 100°C. has been found particularly satisfactory.

The film can be heated by any means adaptable to the particular orientation apparatus used. It has been found that radiant or infrared heaters positioned prior to the nip rolls on both sides of the film provide an effective means of heating the film to the desired embossing temperature range.

The embossing roll should be maintained at least at a temperature within the temperature range to which the film is heated, so as to prevent a temperature drop in the preheated film during embossing. The embossing roll can be heated above this temperature range to further elevate the film surface temperature by the momentary contact of the roll and film at embossing. However, the surface temperature of the film should not be raised more than 40 centigrade degrees above the second order transition temperature. The elevated temperature of the embossing roll causes the film to accept deformation in its plastic state and to retain the plastic deformation due to its effecting heat setting of the surface of the embossed film to prevent relaxation of the embossed pattern. The particular degree of preheating and the embossing roll temperature can vary, so long as the temperature of the bulk of the film at the center of the embossing nip is within the specified temperature range, and the surface or skin temperature at the embossing nip does not exceed 40 centigrade degrees above the second order transition temperature; for polyethylene terephthalate, it is preferred to maintain surface temperature at 110-115°C. because higher temperatures cause excessive crystallinity after embossing. The film surface temperature can be calculated by conventional techniques considering variables such as specific polymers, temperature of the polymer and roll, speed of travel, and heat transfer rate.

The embossing surface can be prepared from any material sufficiently stable to retain its configuration at the required embossing temperatures and pressures. In general, metal surfaced rolls have been found most satisfactory in this application. The surface of the embossing roll can be conveniently prepared by blasting with sand, glass beads, aluminum oxide grit or shot to provide a surface blasted to a surface roughness of about from 30 to 500 microinches (0.76 to 12.7 microns), A.A. The embossing roll surface roughness is measured by conventional techniques, using, for example, a "Gould" surfanalyzer at a 0.03 inch cutoff, according to American Standards (NASI) test B-46.1, 1962. Other techniques for roughening a surface are plasma jet coating, photo etching, electric discharge machining and engraving. An embossing roll having a surface roughness of about from 150 to 250 microinches (3.8 to 6.35 microns), A.A., provides a film product having exceptionally good winding performance in films having a final thickness of about 1.5 mils. A blasted steel surface for the embossing roll having been blasted and chrome plate to a surface roughness of about from 30 to 500 microinches (0.76 to 12.7 microns), A.A., provides a significant

winding improvement and is especially preferred for embossing applications in which two relatively smooth surfaces are required on the finished film product. In general, it is believed that a lower degree of surface roughness on the embossing roll will give a comparable improvement in winding characteristics with thinner gauge films.

In those embodiments of the process of the invention wherein the embossing surface is one of a pair of nip rolls used in the machine direction stretching of the film, the pressure applied to the film is that which is sufficient to maintain a firm grip on the film in order to effect the desired degree of stretching. In general, a pressure along the nip of at least about 50 and preferably about 100 pounds per lineal inch (9 kg/cm and preferably about 18 kg/cm) can be used, although the particular pressure required will vary with the composition of the polymer, the thickness of the film, the rate of travel as well as the degree of stretch. The second of the pair of nip rolls is generally a more resilient material than the embossing roll, such as a silicone rubber. Important characteristics of the second of the two nip rolls include sufficient rigidity to apply the embossing pressure and to impress the pattern to be embossed upon the film surface, preferably 50 to 70 Durometer Shore A hardness. The second roll is generally relatively smooth in order to provide one film surface relatively free from irregularities and asperities for the best possible performance as a magnetic tape base.

In a preferred embodiment of the invention, the second, backup, nip roll is cooled below the embossing temperature range in order to facilitate the rapid cooling of the film surface in contact therewith and to avoid imparting defects to this surface from the backup roll. The cooling of the backup roll can be effected by maintaining the backup roll in contact with an independent roll maintained at a substantially lower temperature.

Immediately after passing through the embossing nip, the embossed film preferably is cooled below the second order transition temperature, and more preferably below 70°C., in order to minimize relaxation of the embossed pattern. The film can be cooled by any cooling means adaptable to the particular stretching and embossing apparatus used to effect the instant process including, for example, a stream of refrigerated air directed toward the embossed film immediately outside of the embossing nip. It is preferred, however, that the embossed film be cooled by passing the film over a quench roll maintained at a temperature substantially below the second order transition temperature of the polymer. Quench roll temperatures of about 15°C. have been found particularly effective in minimizing the relaxation of the embossed pattern.

Thereafter, the film is stretch oriented in the transverse direction according to methods conventionally used in the art. The amount of such stretching will control the ultimate surface characteristics; for example, increasing transverse stretching from 3.4X up to 4.0X results in a 25% reduction in peak-valley height of the ridges on the finished film.

The biaxially oriented films of the invention exhibit one relatively smooth surface and a second surface having a pattern of substantially flat-topped ridges thereon. The ridges represent an elongation of the embossed pattern of protuberances resulting from the transverse orientation of the film after embossing. Since the films are generally oriented in the transverse direction by stretching at least two times their original dimension, substantially round protuberances produce ridges having length-to-width ratios of at least 2:1. In a similar manner post stretching of the biaxially oriented film, however, will reduce the height and also the length-to-width ratio of the ridges so that the ratio will be below 2:1 but greater than 1:1. The ridges should have a peak-to-valley height of at least about 10 microinches (0.25 micron), and up to about 80 microinches (2.03 microns) when embossing rolls are used having a surface roughness of about 500 microinches (12.7 microns), A.A. Preferably, the peak-to-valley height of the ridges is less than about 50 microinches (1.27 microns). Films having been embossed with a 150-microinch (3.8-micron), A.A., surface typically exhibit peak-to-valley ridge heights of 15-35 microinches (0.38-0.89 micron) on the embossed surface. Typical ridges have widths of about from 4 to 10 mils (100 to 250 micron), lengths of about from 10 to 50 mils (250 to 1250 microns) and peak-to-valley heights of about from 10 to 15 microinches (0.25-0.38 micron), i.e. the width of the ridges is greater than their height.

The dual-surfaced films of this invention exhibit exceptionally good winding characteristics. A convenient measure of winding performance is obtained through the use of the Winding Slope of the film as described in German OLS No. 1,961,953 hereby incorporated by reference. The winding slopes exhibited by the subject dual-surfaced films are exceptionally high, typically falling within the range of about 230 to 330. While the reason for the especially high winding slopes is not fully understood, it is believed to be a function of the transverse orientation of the pattern appearing on one surface of the films, the transverse ridges creating passageways for air between the surfaces of

adjoining film layers, thus permitting adjustment between film surfaces on the formed roll. In addition, a minimum peak-to-valley height of the ridges of about 10 microinches (0.25 micron) has been found necessary for good winding performance.

An apparatus which can be used in the process of the instant invention is schematically illustrated in the accompanying drawing, wherein extruded film *10* is stretched in the machine direction by passing first through the nip formed by rolls *11* and *12*, operating at a slow speed, and subsequently passing the film past radiant heaters *13* and *14*, positioned on either side of the film. The heated film is then passed through the nip formed by rolls *15* and *16*, moving at a speed faster than rolls *11* and *12*, thus effecting a machine-direction stretching of the film. The surface of roll *16* may have been prepared by blasting as previously described, to permit embossing one surface of the preheated film. Roll *16* is heated to a temperature of about 95 to 140°C. to effect a permanent deformation of the machine direction oriented film and simultaneously heat set the embossed pattern into the surface of the film. Roll *15* can have a silicone rubber finish and is preferably in contact with cooling roll *17* which is maintained at a temperature of about 15°C. The cooling roll maintains the surface of roll *15* at a temperature lower than the surface of roll *16*. Typical surface temperatures of roll *15* when in contact with cooling roll *17* are about 90°C. An embossing pressure of about 100 pounds per lineal inch (18 kg/cm) is applied at the nip along the heated film at the nip gap.

The quench roll *18* is positioned adjacent roll *15* to quench the embossed surface of the machine direction oriented film and simultaneously remove the embossed film from the surface of the nip roll. The quench roll is generally maintained at a temperature of about 15°C. The film is then oriented in the transverse direction according to conventional techniques.

The process of the instant invention is further illustrated by the following specific example.

Example

A polyethylene terephthalate film is oriented by stretching between two sets of nip rolls moving at different speeds, in an apparatus similar to that illustrated in the drawing. The film is heated by infrared heaters to a temperature of about 100°C.

Nip roll *16* is a steel roll which, having been blasted with "Crystalon" (Registered Trade Mark) 90 mesh aluminum oxide grit, has a maximum surface variation, peak-to-valley, of 200 microinches (5.7 microns) and exhibits a surface roughness of about from 40 to 50 microinches (1.0 to 1.27 microns), A.A. The nip roll is maintained at a surface temperature of 120°C.

The machine direction oriented film, having a thickness of about 3 mils (76 microns), is passed through the nip between rolls *15* and *16* at a pressure of about 100 pounds per lineal inch (18 kg/cm) and a speed of about 68 feet per minute (20.8 meters/minute). The second nip roll *15* is surfaced with a silicone rubber, and is in contact with quench roll *17* to give the nip roll a surface temperature of about 90°C.

After passing through the embossing nip, the film is removed from roll *15* by quench roll *18* maintained at a temperature of about 15°C. After quenching, the pattern replicated onto the embossed surface of the machine direction oriented film exhibits a roughness of about 55 microinches (1.4 microns) in peak-to-valley dimension. The film is then reheated and oriented in the transverse direction to give a final biaxially oriented film product having one smooth surface and one embossed surface characterized by substantially flat-topped transverse ridges having a height of 10 to 15 microinches (0.25-0.38 micron), a width of about 4 mils (100 micron) and a length of about 16 mils (400 micron), i.e. the width:height ratio of the ridges is greater than 100.

The biaxially oriented film has exceptionally good winding characteristics, exhibiting winding slopes of 230 to 330.

WHAT WE CLAIM IS:—

1. Biaxially oriented polyester film adapted by surface roughness for rapid winding into uniform stable rolls characterized by a multiplicity of transverse direction ridges on one surface of the film, said ridges having a length to width ratio of greater than 1:1 and a peak-to-valley height of at least 10 microinches (0.25 micron).

2. A film according to Claim 1 wherein the ratio of length to width is at least about 2:1.

3. A film according to Claim 1 or 2 wherein ridge height is up to 80 microinches (2.03 microns).

4. A film according to Claim 1 or Claim 2 wherein ridge height is 10 to 15 microinches (0.25 to 0.38 micron).

5. A film according to any of Claims 1 to 4 wherein ridge width is 4 to 10 mils and ridge length is 10 to 50 mils.

6. A film according to any of Claims 1 to 5 wherein one surface has ridges and the other surface is smooth.

7. A film according to any of Claims 1 to 6 wherein the polyester is polyethylene terephthalate.

8. A film according to Claim 6 or 7 in the form of magnetic tape.

9. A film according to any of Claims 1 to 8 having a winding slope of 230 to 330.

10. A process for making a film according to any of Claims 1 to 9 wherein protuberances are embossed onto one surface of the film after machine direction stretch orienting and before transverse direction stretch orienting by

(a) heating the film to a temperature range R, between the second order transition temperature and about 40 centigrade degrees above the second order transition temperature,

(b) bringing the film into contact with an embossing surface having a surface roughness of from 30 to 500 microinches (0.76 to 12.7 microns), A.A., and at a pressure of at least about 50 pounds per lineal inch (9.0 kg/cm), the embossing surface being heated to provide a film surface temperature of at least temperature range R, and

(c) converting the protuberances into transverse ridges by transverse direction stretch orienting the film.

11. A process according to Claim 10 wherein, immediately after embossing, the film is quenched to below the second order transition temperature.

12. A process according to Claim 11 wherein quenching is accomplished by passing the film over a quench roll maintained at about 15°C.

13. A process according to any of Claims 10 to 12 wherein embossing is accomplished by providing an embossing surface on a machine direction stretch fast nip roll.

14. A process according to any of Claims 10 to 13 wherein surface roughness of the embossing surface is 150 to 250 microinches (3.8 to 6.35 microns), A.A.

15. A process according to any of Claims 10 to 13 wherein the embossing surface is provided by a roll having a blasted steel surface of surface roughness from 40 to 50 microinches (1.0 to 1.2 microns), A.A.

16. A process according to any of Claims 10 to 15 wherein the embossing surface is heated.

17. A process according to Claim 16 wherein the surface is heated above range R to provide temperature of the bulk of the film, at the middle of the embossing nip, of greater than 20 centigrade degrees higher than the second order transition temperature of the film.

18. A process according to Claim 17 wherein the film is polyethylene terephthalate, said temperature of the bulk of the film is about 100°C., and temperature of the surface of the film is 110 to 115°C.

19. A process according to any of Claims 10 to 18 wherein the embossing surface is provided by one roll of a pair of machine direction stretch fast nip rolls, the other roll of said pair being a resilient smooth surfaced roll of 50 to 70 Durometer Shore A hardness.

20. A process according to Claim 19 wherein said other roll has a surface of silicone rubber.

21. A process according to any of Claims 10 to 20 wherein the film is brought into contact with the embossing surface at a pressure of at least about 100 pounds per lineal inch (18 kg/cm).

22. A process for making biaxially oriented polyester film substantially as hereinbefore described with reference to the accompanying drawing and/or the Example.

23. Biaxially oriented polyester film when made by the process of any one of Claims 10 to 22.

24. Biaxially oriented polyester film substantially as hereinbefore described in the Example.

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1303971 COMPLETE SPECIFICATION

1 SHEET *This drawing is a reproduction of the Original on a reduced scale*